

Radiochemical characterization of mineral waters in the Eastern Black Sea Region, Turkey

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Abstract This study has evaluated the levels of natural radionuclides and chemical components of mineral waters in the Eastern Black Sea Region (Turkey). The mean activity concentrations of ^{226}Ra , ^{232}Th , ^{137}Cs , ^{40}K , gross alpha and gross beta were found as 129, 33, 28, 714, 125 and 170 mBq L^{-1} , respectively. Due to consumption of mineral waters, the radiological impact of them on the inhabitants was calculated by taking the annual intake into account through ingestion of aforementioned radionuclides. The estimated effective doses from mineral water were found to be $13.20 \mu\text{Sv year}^{-1}$ (^{226}Ra), $2.74 \mu\text{Sv year}^{-1}$ (^{232}Th), $0.13 \mu\text{Sv year}^{-1}$ (^{137}Cs) and $1.62 \mu\text{Sv year}^{-1}$ (^{40}K). The overall contribution of these radionuclides to the committed effective dose from a year's consumption of mineral water in the region is therefore estimated to be only 17.69%, which is in

concordance with the recommended WHO value ($100 \mu\text{Sv year}^{-1}$). The chemical analysis results showed that these waters contain Na, Al, P, Cl, K, Ca, V, Mn, Fe, Ni, Cu and Zn elements. These values were evaluated and compared with the internationally verified values. This study provides important information for consumers and authorities because of their internal radiochemical exposure risk from mineral water intake.

Keywords Radioactivity · Gross alpha and beta · Chemical analysis · Mineral water · Turkey

Introduction

Mineral water is water containing minerals or other dissolved substances that alter its taste or give it therapeutic value. Recently, there has been an increasing tendency to replace tap water with mineral water for consumption purposes. According to this trend, the consumption of mineral water has increased rapidly and has become popular among the public. As it is very important for human life, its quality must be carefully and systematically controlled. The reason for this is that there is an increasing health risk due to the dissolved natural radionuclide content of water, predominantly because of the members of the uranium and thorium decay series. Their activity concentrations depend on the origin of the water;

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therefore, they vary considerably. Several physical and geochemical processes might affect the level of radionuclides in underground waters; hence, the consumption of mineral water might result in increasing natural radiation exposure (Jobbagy et al. 2010).

Activity in a radioactive substance which can be ingested or inhaled into the body may produce a damaging biological effect and can create serious health risks. That is to say that the occurrence of radionuclides and heavy metals in water causes human internal exposure due to the decay of radionuclide absorbed by the body through ingestion (Fatima et al. 2007). Hence, contamination of mineral waters with radionuclides and heavy metals present great hazard.

The Eastern Black Sea region consists of deep running water valleys, mountains, steps and broken zones. All streams rising from this land flow into the Black Sea. Economic activities are based on agriculture and animal breeding, so industry has not yet developed sufficiently. According to 2007 census results, the population of the Eastern Black Sea region is approximately 2.6 million. While 1.4 million people live in cities, the rest live in villages. In the region, mineral water has been naturally consumed by the local people, and in the future, it may be bottled and transported to other parts of the country for consumption.

Over the past several years, some results of radioactivity levels have been published in some papers for tap waters (Damla et al. 2006; Cevik et al. 2006) and for natural spring water (Kobyas et al. 2010) in the Eastern Black Sea region, Turkey. However, the detailed information of mineral waters is not available in literature for the Eastern Black Sea region. Therefore, the objective of this

work is to determine the radiochemical characterization in the mineral waters and to estimate the radiological implications on the population using these waters in this region. The results obtained in the present study are compared with internationally recommended values and reported data of other countries.

Materials and methods

Sampling area

In order to evaluate radiochemical characterization of mineral waters in the Eastern Black Sea Region, Turkey, totally 13 mineral water samples were collected from six different provinces for analyses. Figure 1 shows the locations where the samples were collected.

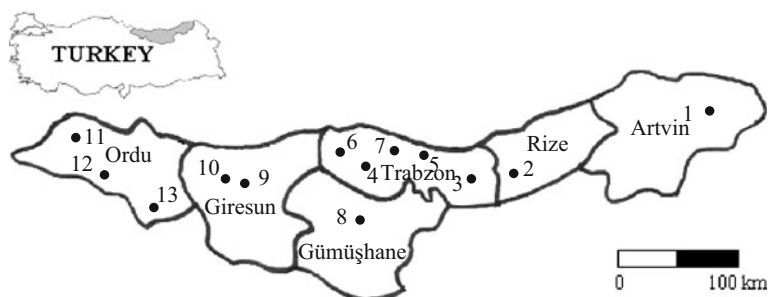
Chemical analysis

Five hundred millilitres of mineral water sample was collected from each of sampling point. The water samples were bottled after adding 1% HCl acid solution. Collected samples were filtered in the laboratory before measurement. An inductively coupled plasma optical emission spectrometer (Spectro Genesis model) was used for Na, Al, P, Cl, K, Ca, V, Mn, Fe, Ni, Cu and Zn determinations. The information of the spectrometer was described in detail in earlier publication (Kobyas et al. 2010).

Determination of gross α and β activity in waters

All the samples were collected in polypropylene bottles of 1.5 L capacity. The bottles were

Fig. 1 Location of sampling sites indicating the Eastern Black Sea, Turkey



cleaned using a modified procedure (Laxen and Harrison 1981). Of the collected water samples, about 250 mL was acidified with dilute of nitric acid to be pH 1 to avoid the collection of organic materials and changes in the state of the ions that were present in the samples. Subsequently, samples were slowly evaporated (avoiding boiling) in a furnace temperature at 60°C down to a 50 mL volume. The residue was transferred quantitatively to a stainless-steel planchet and dried. Then, the sample was allowed to cool down in a desiccator before determining the weight of it. Before measurement, the samples were well dried in a furnace at a temperature of 105°C (Krieger 1975). The counting time was 30,000 s for gross α and β activities.

The instrumentation to count the gross α and gross β activities was an α/β counter of the low background multiple detector type with 10 sample detectors (Berthold LB770). Typical detection limits for a counting time of 500 min were estimated to be 3 mBq L⁻¹ for gross α and 6 mBq L⁻¹ for gross β . The calibrations of the spectrometer were described in detail in earlier publications (Damla et al. 2006, 2009).

Determination of radionuclide concentrations

In order to measure the radionuclide concentrations in mineral waters, various samples were collected in 30-L polypropylene bottles. The bottles were first soaked in 10% HNO₃ for 48 h, followed by de-ionized water for a few minutes and finally rinsed three times with double-distilled, de-ionized water. The collected water samples were acidified with concentrated nitric acid to pH 1 to break down the organic materials and to prevent loss of ions in the sample following binding to the container or precipitation. Subsequently, the samples were slowly evaporated down to a volume of 100 mL in a furnace at 60°C and poured into cylindrical, polyethylene plastic containers (5.5 cm diameter and 5 cm height) for gamma activity analysis. These samples were then left for 30 days to allow ²²⁶Ra and its short-lived progeny to reach radioactive equilibrium.

Gamma spectrometry measurements were performed with a coaxial HPGe detector at a 15% relative efficiency and a resolution of 1.9 keV

at the 1,332 keV gamma of ⁶⁰Co (Canberra, GC 1519 model). The energy calibration and absolute efficiency calibration of the spectrometer were described in detail in earlier publications (Cevik et al. 2008; Damla et al. 2010).

The gamma-ray lines of 352 keV from ²¹⁴Pb and 609 keV from ²¹⁴Bi were used to evaluate the ²²⁶Ra activity concentration (Canet and Jacquemin 1990; Amrani 2002; Ajayi and Owolabi 2008), while 583 keV gamma-rays from ²⁰⁸Tl and 911 keV from ²²⁸Ac were used to determine to the ²³²Th activity concentrations. The activity concentrations of ⁴⁰K and ¹³⁷Cs were determined by using their 1,460 and 661 keV gamma-ray lines, respectively. The specific activity of each sample was then calculated utilizing the following formula:

$$C = \frac{N}{\varepsilon PMt} \text{ (Bq L}^{-1}\text{)} \quad (1)$$

where N is the net counting rate of the gamma ray, ε the detector efficiency of the specific gamma ray, P the absolute transition probability of gamma decay, M the volume of the sample (L) and t is counting time.

Results and discussions

The results of chemical analyses of 13 investigated mineral waters are presented in Table 1. The elemental concentrations are expressed in micrograms per litre. It can be seen from the table that the mean elemental concentrations decrease in the following order Na > Ca > K > Cl > Zn > Al > Mn > V > P > Fe > Ni. The Mn, Ni and Cu value for the waters are in the range 2–559, 1–45 and 5–26 $\mu\text{g L}^{-1}$ with a mean 144, 14 and 9 $\mu\text{g L}^{-1}$, respectively. These results are below the guideline values recommended by the Codex Alimentarius Commission (CAC 1997), the World Health Organization (WHO 2006) and the Turkish natural mineral water regulations (Turkish Official Gazette 2004). The maximum value of Zn for the waters is found in the sampling code 11 (772 $\mu\text{g L}^{-1}$), while the minimum value is measured in the sampling code 8 (4 $\mu\text{g L}^{-1}$). For Al, six of the samples were found to exceed the recommended values by the World Health Organization (WHO 2006). All but one is above the values the

Table 1 Chemical component of the mineral waters ($\mu\text{g L}^{-1}$)

Sampling code	Province	Na	Al	P	Cl	K	Ca	V	Mn	Fe	Ni	Cu	Zn	U ^a	Th ^a
1	Artvin	BDL	15	76	70,350	33,239	11,975	163	16	33	10	12	573	21.2	5.4
2	Rize	11,816	61	130	756	373	8,551	34	60	20	25	5	23	10.2	12.3
3	Trabzon	70,112	22	70	83,780	3,555	20,173	127	2	22	9	6	376	4.8	4.4
4		70,269	24	106	13,632	3,212	30,225	105	559	22	25	6	286	12.7	5.4
5		32,021	19	127	568	1,294	30,217	83	131	19	7	5	194	9.0	17.9
6		28,642	16	83	710	1,142	30,139	155	8	23	9	9	486	10.8	13.5
7		BDL	166	270	987	26,961	12,185	204	205	22	1	9	638	8.4	6.6
8	Gümüşhane	35,464	213	90	873	480	18,149	191	500	19	45	7	4	7.6	BDL
9	Giresun	62,938	153	74	139,515	2,744	14,440	185	2	21	8	8	552	9.2	6.1
10		8,236	24	89	1,065	1,952	30,115	87	236	20	11	13	5	14.1	5.4
11	Ordu	50,872	148	85	8,520	3,137	11,437	239	47	19	8	5	772	8.7	8.1
12		55,558	131	83	1,420	4,115	13,140	104	5	20	7	6	316	7.1	5.4
13		15,521	1,601	73	4,189	3,580	20,180	43	103	21	12	26	13	9.3	5.4
Mean		40,132	199	104	25,105	6,599	19,302	132	144	22	14	9	326	10.3	7.4
Turkish Standards, Turkish Official Gazette (2004)															
WHO standards (2006)															
US EPA (1993)															
Codex Alimentarius Commission (1997)															
BDL below detection limit															
^a Calculated values															

Turkish natural mineral water regulations (Turkish Official Gazette 2004) and United States Environmental Protection Agency (US EPA 1993).

The specific activities of the radionuclides of ^{226}Ra , ^{232}Th , ^{137}Cs and ^{40}K were determined by using gamma-ray spectroscopy and the gross alpha and gross beta activities were using a α/β counter of the low background multiple detector type with 10 sample detectors (Berthold LB770). The results are given in Table 2. The activity concentrations for ^{226}Ra vary from 61 to 267 mBq L^{-1} . The activity level for ^{232}Th in the water samples ranged from <BDL to 73 mBq L^{-1} with a mean of 33 mBq L^{-1} . It should be noted that in all water samples, the concentrations of ^{226}Ra are higher than those of ^{232}Th . This is probably a result of the granite rocks commonly found in the Eastern Black Sea region (Kobya et al. 2010). The high concentrations of ^{226}Ra reflect the fact that radium is more soluble in ground water than its thorium and uranium precursors (Kitto and Sook Kim 2005). ^{226}Ra is a highly radiotoxic radionuclide. When humans ingest radium, 20% is absorbed into the blood stream. The absorbed radium is initially distributed to soft tissues and bones, but its retention is mainly in growing bones (Wrenn et al. 1985). Elemental concentrations of U and Th elements are calculated using ^{226}Ra and ^{232}Th activity concentrations, respectively. Calculated values are presented in Table 1 in units of micrograms per litre. Table 2 also present activity concentration calculated for ^{40}K and ^{137}Cs content in these waters. The mean activity concentration

of ^{40}K is measured as 714 mBq L^{-1} . For the artificial radionuclide with a ^{137}Cs , the mean activity concentration is 28 mBq L^{-1} . It is great probably considered that ^{137}Cs radionuclide measured is because of atmospheric residue of nuclear weapon tests conducted by several countries; however, some of the residual amount may be the result of Chernobyl accident, which occurred in 1986, which significantly affected across the northern parts of Turkey.

The measurement results of gross alpha and gross beta activities for the water samples collected in this study are presented in Table 2. The gross beta activity has a mean of 170 mBq L^{-1} and ranges from 13 to 854 mBq L^{-1} . The gross alpha activity has a mean of 125 mBq L^{-1} and varies between 7 and 898 mBq L^{-1} . The maximum gross alpha and beta activity is from sampling code 11 of Trabzon province and the minimum gross alpha and beta activity is from sampling code 2 of Rize province. Gross alpha and beta activity in all measured mineral water samples is much more below than the guideline value of 1,500 and 2,000 mBq L^{-1} for gross alpha and beta values, respectively, recommended by the Turkish natural mineral water regulations (Turkish Official Gazette 2004).

It was estimated by Alam et al. (1999) that the effective dose that an individual is exposed to due to the intake of natural radionuclides from water as shown below:

$$D = C \times I \times E \quad (2)$$

Table 2 Natural and artificial activity concentrations (in mBq L^{-1}) of mineral water samples

Sampling code	Province	^{226}Ra	^{232}Th	^{137}Cs	^{40}K	Gross α	Gross β
1	Artvin	267	22	25	108	122	609
2	Rize	128	50	28	801	7	13
3	Trabzon	61	18	BDL	BDL	31	56
4		160	22	25	453	326	129
5		114	73	42	453	16	19
6		136	55	BDL	711	9	24
7		106	27	BDL	1,088	898	854
8	Gümüşhane	96	BDL	19	801	43	139
9	Giresun	116	25	24	1,404	22	62
10		178	22	33	388	15	49
11	Ordu	110	33	33	1,358	46	51
12		90	22	29	469	40	58
13		117	22	24	532	50	152
Mean		129	33	28	714	125	170

BDL below detection limit

Table 3 Annual effective doses to adult member of public in the Black Sea Region (Turkey) from intake of radionuclides

Sampling code	Province	^{226}Ra ($\mu\text{Sv year}^{-1}$)	^{232}Th ($\mu\text{Sv year}^{-1}$)	^{137}Cs ($\mu\text{Sv year}^{-1}$)	^{40}K ($\mu\text{Sv year}^{-1}$)	Total dose ($\mu\text{Sv year}^{-1}$)
1	Artvin	27.29	1.85	0.12	0.24	29.50
2	Rize	13.08	4.20	0.13	1.81	19.22
3	Trabzon	6.23	1.51	BDL	BDL	7.74
4		16.35	1.85	0.12	1.03	19.35
5		11.65	6.13	0.20	1.03	19.01
6		13.90	4.62	BDL	1.61	20.13
7		10.83	2.27	BDL	2.46	15.56
8	Gümüşhane	9.81	BDL	0.09	1.81	11.71
9	Giresun	11.86	2.10	0.11	3.18	17.25
10		18.19	1.85	0.16	0.88	21.08
11	Ordu	11.24	2.77	0.16	3.07	17.24
12		9.20	1.85	0.14	1.06	12.25
13		11.96	1.85	0.11	1.20	15.12
Mean		13.20	2.74	0.13	1.62	17.69

BDL below detection limit

where D is the annual effective dose of an individual due to ingestion of radionuclides (Sv year^{-1}), C is the activity concentration of radionuclides in the ingested mineral water (Bq L^{-1}) and I represents the annual intake of mineral water (L year^{-1}). Assuming that every adult drinks 1 L/day, the annual effective doses caused by intake of the ^{226}Ra , ^{232}Th , ^{137}Cs and ^{40}K isotopes were calculated (Chau and Michalec 2009; Kovacs et al. 2004). E is the ingested dose conversion factor for the radionuclide (Sv Bq^{-1}). The dose conversion factors used for the calculation were taken from the WHO publication (WHO 2006) and were equal to 2.8×10^{-4} , 2.3×10^{-4} and $1.3 \times 10^{-5} \text{ mSv Bq}^{-1}$ for ^{226}Ra , ^{232}Th and ^{137}Cs , respectively. And for ^{40}K , intake was calculated using

the conversion factor of $6.2 \times 10^{-6} \text{ mSv Bq}^{-1}$ (Delacroix et al. 2002). The calculated effective doses are given in Table 3. For adults, the range of effective doses due to intake of ^{226}Ra , ^{232}Th , ^{137}Cs and ^{40}K were 6.23–27.29 $\mu\text{Sv year}^{-1}$, BDL–6.13 $\mu\text{Sv year}^{-1}$, BDL–0.20 $\mu\text{Sv year}^{-1}$ and BDL–3.18 $\mu\text{Sv year}^{-1}$, respectively, with means of 13.20, 2.74, 0.13 and 1.62 $\mu\text{Sv year}^{-1}$, respectively. The total doses for the water are in the range 7.74–29.50 $\mu\text{Sv year}^{-1}$ with a mean 17.69 $\mu\text{Sv year}^{-1}$. The total contribution of these radionuclides to the annual effective dose from a year's consumption of mineral water is therefore estimated to be only 17.69% of the WHO value (100 $\mu\text{Sv year}^{-1}$). As far as the measured radionuclides are concerned, the mean dose contribution

Table 4 Comparison of natural radionuclides in mineral waters (in mBq L^{-1}) with other countries

Country	^{226}Ra	^{232}Th	^{40}K	Gross α	Gross β	Reference
Poland	0.8–525		<BDL–10,064	<BDL–938	<BDL–1,784	Chau and Michalec (2009)
Hungary	4–2,940			35–2,600	<8–3,340	Kovacs et al. (2004)
Brazil	<2–220					Godoy et al. (2001)
Algeria	12–42	15–53	400–1,600			Amrani (2002)
Iran	8–61					Shahbazi-Gahrouei and Saeb (2008)
Italy	<10–53			<4–2,778	<25–930	Desideri et al. (2007)
Poland	<10–335		23–9,904			Kozłowska et al. (2007)
Turkey	61–267	<BDL–73	<BDL–1,404	7–898	13–854	Present study

BDL below detection limit

for the analyzed mineral waters is in the following order 74.6% from ^{226}Ra , 15.5% from ^{232}Th , 9.2% from ^{40}K and 0.7% from ^{137}Cs .

The measured mean activity concentration of ^{26}Ra , ^{232}Th , ^{137}Cs , ^{40}K , gross alpha and gross beta in mineral water samples were compared with different countries of the world in Table 4. For ^{226}Ra , the minimum value measured is higher than that of the other countries mentioned in the Table 4; again for ^{226}Ra , the maximum value measured in this study is higher than Brazil (Godoy et al. 2001), Algeria (Amrani 2002), Iran (Shahbazi-Gahrouei and Saeb 2008) and Italy (Desideri et al. 2007); however, it is lower than Poland (Chau and Michalec 2009), Hungary (Kovacs et al. 2004) and Poland (Kozłowska et al. 2007). While the maximum value found in this study for ^{232}Th is higher than that of Algeria, ^{40}K , gross alpha and gross beta concentrations are generally lower than the literature.

Conclusions

Radiochemical characterization of thirteen mineral waters was performed. The activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs radionuclides in the mineral waters of the Eastern Black Sea region of Turkey were measured using gamma-ray spectrometry. The mean specific activities of the radionuclides in the samples were 129, 33, 28 and 714 mBq L⁻¹, respectively. The computed overall effective dose for ingestion of the radionuclides contained in mineral waters (17.69 $\mu\text{Sv year}^{-1}$) is insignificant compared with the value of 100 $\mu\text{Sv year}^{-1}$ recommended by WHO. The gross alpha and beta activities were using a α/β counter of the low background multiple detector type with 10 sample detectors. Gross alpha and beta activities ranged from 7 to 898 and from 13 to 854 mBq L⁻¹, respectively. The radionuclide concentrations in natural mineral waters in the Eastern Black Sea region are also relatively low and satisfy the WHO regulations for drinking water consumption. In water samples, the activity concentrations of the ^{137}Cs are generally detected. This can be attributed to the effects of the ^{137}Cs concentration that contaminated the Eastern Black Sea Region after the Chernobyl

nuclear power plant accident which happened in 1986. Concentrations of elements in waters were also measured by inductively coupled plasma-optical emission spectrometry. Because some individuals drink a lot of mineral water of their own vicinity, its quality assessment is of primary importance. This study has demonstrated that for the investigated elements the mineral water sources in the Eastern Black Sea region have good chemical quality and meet the WHO, CAC, US EPA and Turkish Official Gazette guidelines. Results obtained from these data may provide setting the national guidelines and safety standards for the permissible concentrations of natural radionuclides in the mineral water. Moreover, it may be useful in forensic studies by establishing background values of radionuclides in the studied area.

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